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Concept of Phase Transition Based on Elastic Systematics

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Abstract

The use of elastic constants systematics to describe fundamental properties of engineering materials has made materials science education and its related subjects increasingly important not only for manufacturing engineers but also for mankind at large. In this chapter, we present actual scaling of phase transition-driven considerations, such as martensitic transformation and transformable shape memory formation via elastic constant systematics. The scaling in terms of the simple and polycrystals mechanical stability criteria based on the elastic moduli and an acoustic anisotropy is in good agreement with novel experimental data from the literatures, and further, a long-standing concern in predicting polycrystalline elastic constants was considered beyond the commonly encountered criteria.

Keywords: elastic, elastic modulus, martensitic transformation, shape memory effect, elastic constant, ductility criterion, mechanical properties

1. Introduction

The ingenuity and the art required to tailor precisely the desired physical and structural properties in materials have been the main goal of the material scientists and engineers. Elastic response (i.e. elastic constant) to an applied load is one of such basic properties of all solids and originates from the distortion of atomic bonds. Simply put, elastic constants are a reflection of the fundamental thermodynamic properties that take place in the crystal lattice of solids. Complementary to this, the otherwise inaccessible essential information can be revealed from their temperature and stress dependencies of these important constants. For instance, the crystal structures of the three long periods of transition elements change more or less systematically from hcp through bcc to fcc as their group numbers increase from IV to VIII as does their elastic properties. Thus, the knowledge of microscopic elasticity can provide a fruitful ground for the exploration of the material behaviour yet uncommon to our knowledge about the relationship between crystal structure and bonding.

The earliest foundation of elastic theory dates back to seventeenth century (around 1821), when Navier first gave the equation for the equilibrium and motion of elastic solids [1], but modern foundation of microscopic elastic theory was established by the work of Born and Huang [2], followed by other excellent treatments [3]. It is well known that crystalline solids are by no means ideal and invariably contain some lattice defects such as vacancies, solute atoms or some extent of disorder. These point defects strongly affect almost all properties of

materials, including elastic behaviours. In effect, the early investigators of these phenomena were motivated by the response of naturally occurring anisotropic materials such as wood and other crystalline solids. On that premise, of interest here is the relationship between crystal structure and elastic properties, mainly because of the important information they provide about nature of binding forces in solids.

Over the past three decades, elastic constants of some simple crystals have been a subject of numerous researches and have been investigated both theoretically and experimentally. Some of the outcomes have revealed that fundamental elastic properties of a martensitic crystal are fully determined by the elastic constants C_{ij} . All macroscopic elastic moduli (Young's and shear modulus, Poisson ratio, etc.) can be derived from the C_{ij} at least within certain upper and lower bounds [4]. There is considerable evidence that the magnitude of $C' = (C_{11} - C_{12})/2$ elastic shear modulus in metallic bcc structures is closely related to the occurrence of martensitic phase transformations and is thus a useful parameter for estimating bcc structures [5]. Similarly, whether a structural material shows plastic flow or brittle fracture on loading is of clear practical significance. Brittleness in polycrystalline metals can be intrinsic or induced. The basic question is: Do these two general properties (i.e. phase stability and elastic properties) of crystals correlate to each other?

2. Analytical criterion of elastic constants of perfect crystals

The elastic properties are among the most important physical properties of materials and the importance of studying elastic properties of materials cannot be overemphasised. The knowledge of elastic properties is essential for both structural design and experimental mechanics [6]. It also enables the assessment of the sufficiency of strength, stiffness and stability of newly developed materials. Although the crystals are assumed to free from lattice imperfections and difficult to produce, their study had always been the building block for a better understanding of the behaviour of bulk materials. Usually, the determination of elastic properties of crystalline solids is based on its single or perfect crystal configuration under special loading conditions. The elastic moduli are the material constants that connect stress with strain and are therefore crucial to engineering applications. A crystal subjected to external load undergoes dimensional change. If the external load is a stress tensor denoted by σ_{ij} , then the deformation per unit length in three-dimensional space, can be described by a strain tensor, e_{ij} . Within the elastic limit or for sufficiently small deformations, the stress tensor is a linear function of the strain tensor and the generalised delta notation of Hooke's law can be used to express the relationship between these two quantities [7] as:

$$\sigma_{ij} = C_{ijkl}e_{kl} \quad (1)$$

where C_{ijkl} is the proportionality constant that characterises the crystal's resistance to elastic shape change; often referred to as the elastic coefficients or elastic constants or elastic moduli or stress-strain coefficients [8].

The inverse relation between the strain and the stress can be determined by taking the inverse of stress-strain relation to get:

$$e_{ij} = S_{ijkl}\sigma_{kl} \quad (2)$$

Here, S_{ijkl} represents the elastic compliance of the crystal. From symmetry or equilibrium principles, the state of stress in an elastic body can be approximated by six independent stress and strain components. And as such the stress and strain components in Eq. (1) can be expressed in three orthogonal axes as:

$$\begin{aligned}\sigma_x &= C_{11}e_{xx} + C_{12}e_{yy} + C_{13}e_{zz} + C_{14}e_{yz} + C_{15}e_{zx} + C_{16}e_{xy} \\ \sigma_y &= C_{21}e_{xx} + C_{22}e_{yy} + C_{23}e_{zz} + C_{24}e_{yz} + C_{25}e_{zx} + C_{26}e_{xy} \\ \sigma_z &= C_{31}e_{xx} + C_{32}e_{yy} + C_{33}e_{zz} + C_{34}e_{yz} + C_{35}e_{zx} + C_{36}e_{xy} \\ \tau_{yz} &= C_{41}e_{xx} + C_{42}e_{yy} + C_{43}e_{zz} + C_{44}e_{yz} + C_{45}e_{zx} + C_{46}e_{xy} \\ \tau_{zx} &= C_{51}e_{xx} + C_{52}e_{yy} + C_{53}e_{zz} + C_{54}e_{yz} + C_{55}e_{zx} + C_{56}e_{xy} \\ \tau_{xy} &= C_{61}e_{xx} + C_{62}e_{yy} + C_{63}e_{zz} + C_{64}e_{yz} + C_{65}e_{zx} + C_{66}e_{xy}\end{aligned}\quad (3)$$

Here, e_{xx} , e_{yy} and e_{zz} are tensile strains, e_{xy} , e_{yz} and e_{zx} are shear strains. The experimental values of elastic constants, C_{ijkl} , were originally determined by considering the response of crystals to small strains or unstressed lattice using Eq. (1). Beyond using Eq. (1) based on measured stress-strain relations, there are now methods of determining elastic constants from the first principles often referred to as ab initio methods. There are many methods of evaluating elastic coefficients such as the one based on expanding the internal strain energy of the crystal [7]. Thus, we may write as Eq. (4),

$$U = U_0 + V_0 \sum \sigma_i e_i + \frac{1}{2} V \sum_i \sum_j C_{ij} e_i e_j + \dots \quad (4)$$

where U is the energy of the crystal, is a quadratic function of the strains, in the approximation of Hooke's law (recall the expression for the energy of a stretched spring). V_0 is its equilibrium volume and e denotes an elastic strain. If the material is a crystal, the number of independent elastic constants is reduced further depending on the crystal system.

Elastic coefficients and elastic moduli have significant effect of mechanical response of crystals. Elastic constants, C_{ij} (C_{11} , C_{12} , C_{44}) and elastic moduli such as bulk modulus (B), shear modulus (G), Young's modulus (E) influence mechanical response of crystals. For instance, the bulk modulus (B) is associated with the hardness of materials which is of extreme importance in high-temperature and pressure applications, while elastic constants could provide essential information about bonding between adjacent atomic planes, anisotropic character of bonding and structural stability [7]. By far, the most widely reported elastic properties are E , G and B , corresponding to tensile, shear and hydrostatic loading, respectively. Since B signifies the compressibility of a substance, it can be calculated from the partial derivative of volume (V) and pressure (P) at constant temperature (T), as per Eq. (5).

$$B = (\delta V / \delta P)_T \quad (5)$$

It is worth pointing out that other definitions of elastic constant are possible. Elastic modulus E measures the resistance to a change in atomic separation distance within the plane of the bond and so can be determined from the linear portion of the interatomic potential. G quantifies the resistance to shear loading and B , since it corresponds to a volumetric dilatation, is dependent on the electronic properties of

a solid, i.e. the compressibility of the electron gas. Elastic moduli are therefore controlled by interatomic interactions and so may be considered a fundamental property of condensed matter. By excitation of longitudinal and transverse phone modes, E and G can, respectively, be calculated if the density (ρ) of the material is known. This is done via an ultrasonic probe which emits and measures the longitudinal (v_l) and transverse (v_t) sound wave velocities, from which E and G can be calculated via Eqs. (6) and (7):

$$E = \rho v_l^2 \quad (6)$$

$$G = \rho v_t^2 \quad (7)$$

E , G and B can also be calculated from C_{ij} elastic constants. For a material with cubic structure, the number of C_{ij} in the elastic tensor can be reduced from 36 to just 9, due to $C_{ij} = C_{ji}$ and there being strong symmetry in a cubic lattice. The resulting relevant C_{ij} are C_{11} , C_{12} and C_{44} .

$$C_{12} = B + \frac{4G}{3} \quad (8)$$

$$C_{11} = 3B - \frac{C_{12}}{2} \quad (9)$$

$$C_{11} = G \quad (10)$$

$$C' = C_{11} - \frac{C_{12}}{2} \quad (11)$$

The tetragonal shear modulus, C , corresponds to a specific phonon vibration mode in the atomic structure, and is thus directional in nature. In comparison, B is non-directional as it relates to a volumetric effect.

$$B = \frac{C_{11} + 2C_{12}}{3} \quad (12)$$

$$G = \frac{C_{11} - C_{12}}{3} \quad (13)$$

3. Elastic and lattice stability criteria

3.1 Lattice stability in perfect crystal

Elastic properties of a material are very important because they check the mechanical stability, ductile or brittle behaviour based on the analysis of elastic constants, C_{ij} , bulk modulus B and shear modulus G . For example, the bulk modulus measures the resistance of the volume variation in a solid and provides an estimation of the elastic response of the materials under hydrostatic pressure. The shear modulus describes the resistance of a material to shape change.

The fundamental understanding of the conditions of mechanical stability of unstressed crystal structure was laid by the work of Max-Born and co-authors in the 1940s [3], and consolidated later in 1954 [3]. This and other text books gave the generic requirements for elastic stability of crystal lattices in terms of elastic constants [3] and offers simplified equivalents of the generic conditions for some high-symmetry classes. The general stability condition can be stated by considering the second-order elastic matrix and the elastic energy of the crystal

deformed homogeneously by infinitesimal strain as shown in Eqs. (14) and (15) [3], respectively:

$$C_{ij} = \frac{1}{V_0} \left(\frac{\partial^2 U}{\partial^2 e_i \partial^2 e_j} \right) \quad (14)$$

$$U = \frac{1}{2} V_0 \sum_{i,j=1}^6 C_{ij} e_i e_j + O(e^3) \quad (15)$$

where U is the elastic energy, V_0 is the volume of unstressed sample, C_{ij} ($i, j = 1-6$) is the elastic constant and e_i and e_j are the applied strains [2]. In Eq. (15), $O(e^3)$ denotes the terms of numerical error in the order e^3 or higher. A crystal lattice is dynamically said to be stable only if elastic energy U is positive for any small deformation [9], which implies that principal minors of the determinant with elements C_{ij} are all positive [3].

Most real materials (cubic and non-cubic polycrystalline structures) have some types of symmetry, which further reduces the required number of independent elastic moduli. In the case of cubic systems, such as bcc, fcc, NaCl type, or CsCl type) structures, in particular, number of independent elastic moduli is reduced from 36 to 9, as $C_{ij} = C_{ji}$ and there being strong symmetry in the two lattices. Therefore, the conditions for stability reduced to a very simple form using three different elastic constants: C_{11} , C_{22} and C_{44} . The mechanical stability criteria are given by [10]:

$$\begin{aligned} C_{11} - |C_{12}| > 0 \\ C_{11} + 2 C_{12} > 0 \\ C_{44} > 0 \\ C_{12} > C_{11} \end{aligned} \quad (16)$$

The condition when $B < 0$ is referred to as spinodal instability.

Although hexagonal and tetragonal systems have the same form for the elastic matrix, the hexagonal has five, while tetragonal has six independent elastic constants. By direct calculation of the Eigen values of the stiffness matrix, according to [11], four conditions can be derived for elastic stability in both classes:

$$\begin{aligned} C_{11} > |C_{12}|; 2C_{13}^2 < C_{33}(C_{11} + C_{12}) \\ C_{44} > 0; C_{66} > 0 \end{aligned} \quad (17)$$

Similarly, for the orthorhombic system, there are nine independent elastic constants: C_{11} , C_{22} , C_{33} , C_{44} , C_{12} , C_{55} , C_{66} , C_{23} and C_{13} . The mechanical stability of the structure at each concentration can be judged by calculated elastic stiffness. According to Born's criteria [3], the requirement of mechanical stability in an orthorhombic system leads to the following equations [12].

$$\begin{aligned} C_{11} > 0; C_{22} > 0; C_{44} > 0; C_{33} > 0; C_{55} > 0; C_{66} > 0; \\ C_{11} + C_{22} > 2 C_{12}; C_{11} + C_{33} > 2 C_{13}; \\ C_{11} + C_{22} C_{33} + 2 C_{12} + 2 C_{23} + 2 C_{13} > 0; \end{aligned} \quad (18)$$

Further to this, conditions for stability for some high-symmetry crystal classes have been studied. However, there is still some confusion about the form of stability criteria for other crystal systems and classes [8].

A crystal lattice is said to be stable in the absence of external load (unstressed condition) and in the harmonic approximation [13] if and only if it has both dynamic and elastic stability. Dynamic stability implies that its phonon modes have positive frequencies for all wave vectors, while its elastic stability is dependent on elastic energy given by Eq. (15) being always positive ($U > 0, \forall \epsilon \neq 0$). Elastic stability criterion is mathematically equivalent to the following necessary and sufficient conditions: the elastic matrix C is definite exactly positive and all Eigen values of matrix C are positive; all the leading principal and arbitrary minors of matrix C are all positive. The closed form expressions for necessary and sufficient elastic stability criteria for other crystal lattices have been studied. While the stability criterion is linear for some crystal lattices, it is quadratic and even polynomial for others. Thus, the mechanical stability of a crystal is combination of the elastic constant and Born's stability criteria. The elastic constant of a stable crystal must satisfy the Born's criteria to prove its mechanical stability.

3.2 Relative stability of polycrystalline materials

In the case of multi-phase stability, multi-phase composites can be obtained based on multiple scattering theory. For example, polycrystalline materials consisting of two phases, namely cubic and orthorhombic phases can be obtained by homogenising the integral elastic response of the multi-phase polycrystalline samples, following the effective medium approach originally applied by Zeller and Dederichs [13] to determine elastic properties of single-phase polycrystals with cubic symmetry. This type of concept was generalised by Middya and Basu [14] and further extended by Middya [15] and by Raabe et al. [16] to multi-phase composites to determine: (i) the elastic single constants and (ii) the volume fraction of the components within a self-consistent T -matrix solution for the effective medium elastic properties of hexagonal, and orthorhombic polycrystals.

The subset of supercells or cubic and orthorhombic symmetries consisting of three (C_{11}, C_{12}, C_{44}) and nine ($C_{11}, C_{12}, C_{13}, C_{22}, C_{23}, C_{33}, C_{44}, C_{55}, C_{66}$) elastic constants, respectively, was calculated by employing the methodology explained in [16–18] for the elastic properties of the multi-component alloys. This can be viewed as a macroscopic homogeneous effective medium consisting of microscopic fluctuations and characterised by an effective stiffness of C_{ijkl} defined by:

$$\langle \sigma_{ij}(\mathbf{r}) \rangle = C_{ijkl} \langle \epsilon_{kl}(\mathbf{r}) \rangle \quad (19)$$

Here, C_{ijkl} is the local elastic constant tensor with $\langle \sigma_{ij}(\mathbf{r}) \rangle$ and $\langle \epsilon_{kl}(\mathbf{r}) \rangle$ as the local stress and strain field at a point \mathbf{r} , respectively, and the angular brackets denoting ensemble averages. A repeated index implies the usual summation convention. The effective stiffness of C_{ijkl} is defined by:

$$\langle \sigma_{ij}(\mathbf{r}) \rangle = C_{ijkl}^* \langle \epsilon_{kl}(\mathbf{r}) \rangle \quad (20)$$

Since the aggregate represents a body in equilibrium, $\sigma_{ij}(\mathbf{r})|_j = 0$, where $|_j = \partial/\partial r_j$ and the local elastic constant tensor can now be decomposed into an arbitrary constants part (C_{ijkl}^o) and a fluctuating part— $\delta C(\mathbf{r})$.

$$C_{ijkl}(\mathbf{r}) = C_{ijkl}^o + \delta C_{ijkl}(\mathbf{r}) \quad (21)$$

As shown in [16], an integral part of Eq. (19) is the interactive equivalent solution representing the resulting local strain ϵ distribution (in a short notation) as:

$$\epsilon = \epsilon^0 + GT\epsilon^0, \quad (22)$$

Here, ϵ^0 and G are the strain and modified Green's function of the medium defined by C^0 , and the T-matrix given by:

$$T = \delta C(1 - G\delta C)^{-1} \quad (23)$$

Here, I is equivalent to the unit tensor. Combining Eqs. (21) and (22), we get:

$$C^* = C^0 + \langle T \rangle / (1 + \langle GT \rangle)^{-1} \quad (24)$$

Although Eq. (21) constitutes an exact solution for C^* , finding the exact solution of $\langle T \rangle$ and $\langle GT \rangle$ for realistic cases is impossible. By neglecting the intergranular scattering that may occur in some cases in the form of a grain-to-grain position-orientation correlation function however, the T-matrix can be written in terms of single-grain T-matrix (t_α) for each grain α

$$T \approx \sum_{\alpha} t_{\alpha} = \tau. \quad (25)$$

where

$$t_{\alpha} = \delta C_{\alpha} + \delta C_{\alpha} G t_{\alpha} = \delta C_{\alpha} (1 - G \delta C_{\alpha})^{-1} \quad (26)$$

$$\sum_{\alpha} \delta C_{\alpha} = \delta C = C - C^0 \quad (27)$$

Inserting Eq. (21) into (22) leads to:

$$C^* = C^0 + \langle \tau \rangle (1 + \langle G\tau \rangle)^{-1} \quad (28)$$

For single-phase polycrystal, the self-consistent solution of Eq. (11) can be obtained by choosing a C^* that satisfies:

$$\langle \tau \rangle = 0 \quad (29)$$

For a multi-phase polycrystals, a solution to Eq. (4) can be found by evaluating the volume fraction and τ of each phase i (u^2 and τ^2), respectively [19], via:

$$\left\langle \sum_i v^2 \tau^2 \right\rangle = 0 \quad (30)$$

The application of the method to both single-phase aggregates and multi-phase composites is relevant to many multi-component alloys. For a single-phase polycrystal with cubic symmetry [16, 20] to the following expression for B^* and $\mu^* : B^* = B^0$

$$8\mu^{*3} + (9B_0 + 4C''')\mu^{*2} - 3C_{44}(B_0 + 4C'')\mu^* - 6B_0C_{44}C'' = 0 \quad (31)$$

In Eq. (31), three independent single-crystal elastic constants (C_{11} , C_{12} , C_{44}) define the single-crystal bulk modulus $B^0 = (C_{11}^0 + 2C_{12}^0)/3$, the tetragonal shear modulus $C' = (C_{11} - C_{12})/2$ and trigonal shear modulus, C_{44} , $\mu^* = C_{44}^* = \mu^0 + \frac{\langle \tau_{44} \rangle}{1 + G_{44} \tau_{44}}$.

Here, C_{44}^* is the homogenised bulk modulus. The details of the equation for calculating the elastic constants of polycrystals alloy with hexagonal symmetry have been explained elsewhere by [20], and the details here concern polycrystals with orthorhombic symmetry. Eqs. (29) and (30) are reduced to a set of coupled equations for B^* and μ^* :

$$0 = 9(K_V - B^*) + 2\beta(d - c + e) + 3\beta^2\Delta' \quad (32)$$

$$0 = \frac{a - b + \beta(2d - 2c - e) + 3\gamma(d - c + e) + \eta\beta\Delta'}{1 - \alpha\beta - 9\gamma(k_v - B_0) + \beta(\beta + 2\gamma)(c - d) - 2e\beta\gamma - \frac{1}{3}\eta\beta^2\Delta''} + 3\left(\frac{C_{44} - \mu^0}{1 - 2\beta(C_{44} - \mu^0)} + \frac{C_{55} - \mu^0}{1 - 2\beta(C_{55} - \mu^0)} + \frac{C_{66} - \mu^0}{1 - 2\beta(C_{66} - \mu^0)}\right) \quad (33)$$

where

$$9K_V = C_{11} + C_{22} + C_{33} + 2(C_{12} + C_{13} + C_{23}), \quad (34)$$

$$B = \frac{1}{3}(C_{11} + 2C_{12})\mu^* = C_{44} \quad (35)$$

$$\gamma = \frac{1}{9}(\eta - 3\beta) \quad (36)$$

$$a = \delta C_{11} + \delta C_{22} + \delta C_{33}; b = \delta C_{12} + \delta C_{13} + \delta C_{23} \quad (37)$$

$$c = \delta C_{11}\delta C_{22} + \delta C_{11}\delta C_{33} + \delta C_{22}\delta C_{33}; d = \delta C_{12}^2 + \delta C_{13}^2 + \delta C_{23}^2 \quad (38)$$

$$e = \delta C_{12}\delta C_{13} + \delta C_{12}\delta C_{23} + \delta C_{13}\delta C_{23} - \delta C_{11}\delta C_{23} - \delta C_{22}\delta C_{13} - \delta C_{33}\delta C_{12} \quad (39)$$

$$\Delta' = \delta C_{11}\delta C_{22}\delta C_{33} + 2\delta C_{12}\delta C_{13}\delta C_{23} - \delta C_{11}\delta C_{23}^2 - \delta C_{22}\delta C_{13}^2 - \delta C_{33}\delta C_{12}^2 \quad (40)$$

$$\delta C_{11} = C_{11} - C_{11}^0 = C_{11} - K^0 - \frac{4}{3}\mu^0; \delta C_{22} = C_{22} - C_{22}^0 = C_{22} - K^0 - \frac{4}{3}\mu^0 \quad (41)$$

$$\delta C_{33} = C_{33} - K^0 - \frac{4}{3}\mu^0; \delta C_{12} = C_{12} - C_{12}^0 = C_{12} - K^0 + \frac{4}{3}\mu^0 \quad (42)$$

$$\delta C_{13} = C_{13} - K^0 + \frac{4}{3}\mu^0; \delta C_{23} = C_{23} - K^0 + \frac{4}{3}\mu^0 \quad (43)$$

$$\beta = \frac{-3(B^* + 2\mu^*)}{5\mu^*(3B^* + 4\mu^*)}, \quad (44)$$

$$\eta/3 = -1/3B^* + 4\mu^*, \quad (45)$$

$$C_{66} = (1/2)(C_{11} - C_{12}) \quad (46)$$

and orthorhombic symmetry has nine of the single crystal elastic constants, namely: C_{11} , C_{22} , C_{33} , C_{44} , C_{55} , C_{66} , C_{12} , C_{23} and C_{13} .

The elastic constants of a multi-phase polycrystals were determined directly by coupling Eq. (13) for τ_{44} and the $(\tau_{11} + 2\tau_{12})$ components of the T-matrix. For materials with cubic symmetry, the equation is defined as:

$$5\tau_{44} = \left(\frac{1}{C_{11} - C_{12} - \tilde{G}^*}\right)^{-1} + 3\left(\frac{1}{C_{44} - \tilde{G}^*}\right)^{-1} \quad (47)$$

$$\tau_{11} + 2\tau_{12} = \frac{3(C_{11} + 2C_{12}) - 9\tilde{B}^*}{3 - (C_{11} + 2C_{12}) - 3\tilde{B}^*} \quad (48)$$

This is where β is defined in Eq. (31) with \tilde{G}^* and \tilde{B}^* replacing G^* and B^* . For materials with orthorhombic symmetry, the equation reads:

$$15\tau_{44} = \frac{a - b + \beta(2d - 2c - e) + 3\gamma(d - c + e) + v\beta\Delta'}{1 - \alpha\beta - 9\gamma(K_v - \tilde{B}_0) + \beta(\beta + 2\gamma)(c - d) - 2e\beta\gamma - \frac{1}{3}v\beta^2\Delta''} + 3 \left(\frac{C_{44} - \tilde{G}^O}{1 - 2k(C_{44} - \tilde{G}^O)} + \frac{C_{55} - \tilde{G}^O}{1 - 2k(C_{55} - \tilde{G}^O)} + \frac{C_{66} - \tilde{\mu}^O}{1 - 2\beta(C_{66} - \tilde{G}^O)} \right) \quad (49)$$

$$\tau_{11} + 2\tau_{12} = \frac{9(K_v - \tilde{B}^0) + 2\beta(d - c + e) + 3\beta^2\Delta'}{3 \left[1 - \alpha\beta - 9\gamma(K_v - \tilde{B}^0) + \beta(\beta + 2\gamma)(c - d) - 2e\beta\gamma - \frac{1}{3}\eta\beta^2\Delta' \right]} \quad (50)$$

Here, β is defined in Eq. (28), η is defined in Eq. (29), and Δ' in Eq. (23). Here, again G^* and \tilde{B}^* replaces G^* and B^* in the equations for β , v and Δ' . As soon as G^* and \tilde{B}^* have been determined, the homogenised Young's modulus $(\tilde{E})^*$ and Poisson's ratio $(v)^*$ for (an elastically isotropic) polycrystal can be determined using standard elasticity relationships. The homogenised polycrystalline Young's modulus is calculated using:

$$E^* = \frac{9\tilde{B}^* G^*}{3\tilde{B}^* + G^*} \quad (51)$$

$$G^* = \frac{3\tilde{E}^* \tilde{B}^*}{9\tilde{B}^* - \tilde{E}^*} \quad (52)$$

4. Correlation of elastic constants with properties of polycrystalline materials

In many problem relating to polycrystalline or anisotropic materials, it is customary to make use of the properties in an elastically isotropic materials. Most of the common metals and engineering alloys, however, exhibit a marked degree of anisotropy in their single-crystal elastic behaviour and it is therefore more desirable to obtain same on the bases of anisotropic elastic property. The fundamental factors determining the intrinsic plasticity or brittleness behaviour in solids have great link with interatomic potentials, for instance, there is a correlation with the ratio of the elastic shear modulus μ to the bulk modulus B . It is evident, elastic moduli show trends with a range of properties, including hardness, yield strength, toughness and fragility [21, 22]. In this section, for limitation of space, we will, in particular, consider elastic aspect of polycrystals materials with respect to their dependency on specific crystal structure.

4.1 Elasticity and ductility criteria

Strength and ductility have always been one of the crucial issues to study for metal materials. The tendency of materials to be ductile or brittle is being predicted using models based on elastic constants. Some of these include that of Pugh criterion [23] and Cauchy pressure as defined by Pettifor [24]. Pugh proposed an empirical relationship between the plasticity and fracture properties showing the ratio G/B indicates the intrinsic ability of a crystalline metal to resist fracture and deform plastically [25]. This represents a competition between plasticity and fracture considering that B and G represent resistance to fracture and plastic deformation, respectively. Thus, the force required to propagate a dislocation is proportional

to Gb where b is the Burgers vector. This implies that a material with high value of the ratio tends to be brittle (fracture is easier and plasticity is much less), while a low value indicates ductility (plasticity is easier and fracture is not). Fracture strength is also proportional to Ba (a , is the lattice constant) since B is related to surface energy, which indicates brittle fracture strength.

These empirical observations implicate G/B as explaining well brittle or tough behaviour [19, 26]. Pugh's criterion is the most widely used model to predict plastic behaviour of materials [27]. Since yield strength and fracture stress scale with shear modulus and elastic constant, respectively, the Pugh's ratio determines the likelihood of material's failure. If the effect of crystal structure is neglected, high value of Pugh's ratio indicates that a material is prone to brittle failure, while low value of G/B implies ductile failure. The large data on polycrystalline pure metals collected by Pugh [2], when he provided a qualitative ranking from ductile (e.g. Ag, Au, Cd, Cu) to brittle (e.g. Be, Ir) behaviour as G/B increases. For cubic close-packed (ccp) metals, the critical ratio $(G/B)_{crit}$ dividing the two regimes is in the range 0.43–0.56, and for hexagonal close-packed metals, it is 0.60–0.63. Cottrell [28] has estimated $(G/B)_{crit}$ for transgranular fracture from measured surface energies: 0.32–0.57 for ccp metals and 0.35–0.68 for body-centred cubic metals. The spread in values for each structure type largely indicates the interrelationship between crystal structure and elastic constant. Each structure type, however, includes metals with widely differing degrees of elastic anisotropy. Detailed analysis requires knowledge of the relevant elastic constants.

On the other hand, the Cauchy pressure ductility criterion is associated with elastic constants of single cubic crystals such as C_{12} – C_{44} and is useful in describing the nature of bonding in a material [27]. When a material has high resistance to bond bending as found in covalently bonded solids, it will have a negative Cauchy pressure ($C_{44} > C_{12}$). This is in contrast with materials with metallic bonding which exhibit positive Cauchy pressure. When compared with Pugh's ductility criterion, ductile and brittle behaviours are considered to be indicated by a positive and a negative Cauchy pressure, respectively. Although Pugh's and Cauchy pressure criteria are adjured to be based on easily measurable properties of materials such as elastic constants, they do not give the critical value dividing brittle and ductile materials. It is proven in certain materials, including metallic glasses and composites, which religiously respect this dividing line [21]. The behaviour is shown graphically in **Figure 1**. A summary of the correlation between C_{12} – C_{44} and $(G/B)_{crit}$

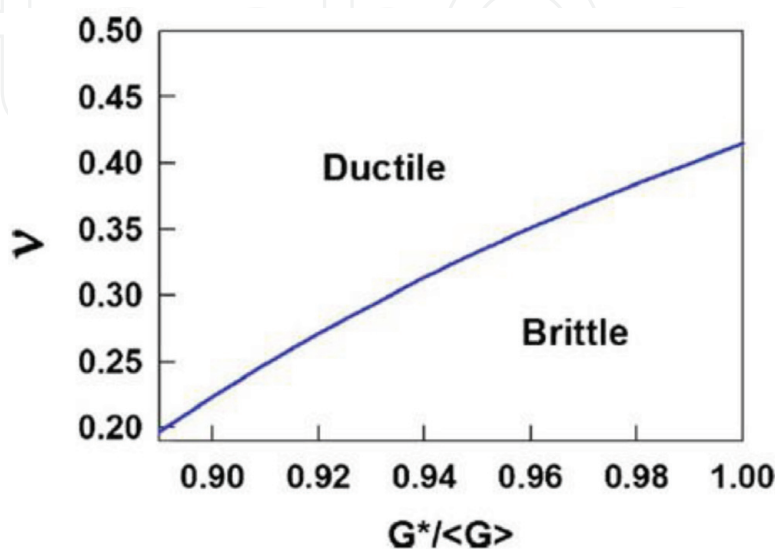


Figure 1.

Ductile and brittle phase fields in metallic glasses, where G^ is the local modulus and ' G ' the global modulus. Decreasing the fraction of low G sites reduces the need for a globally low ν (culled from [28]).*

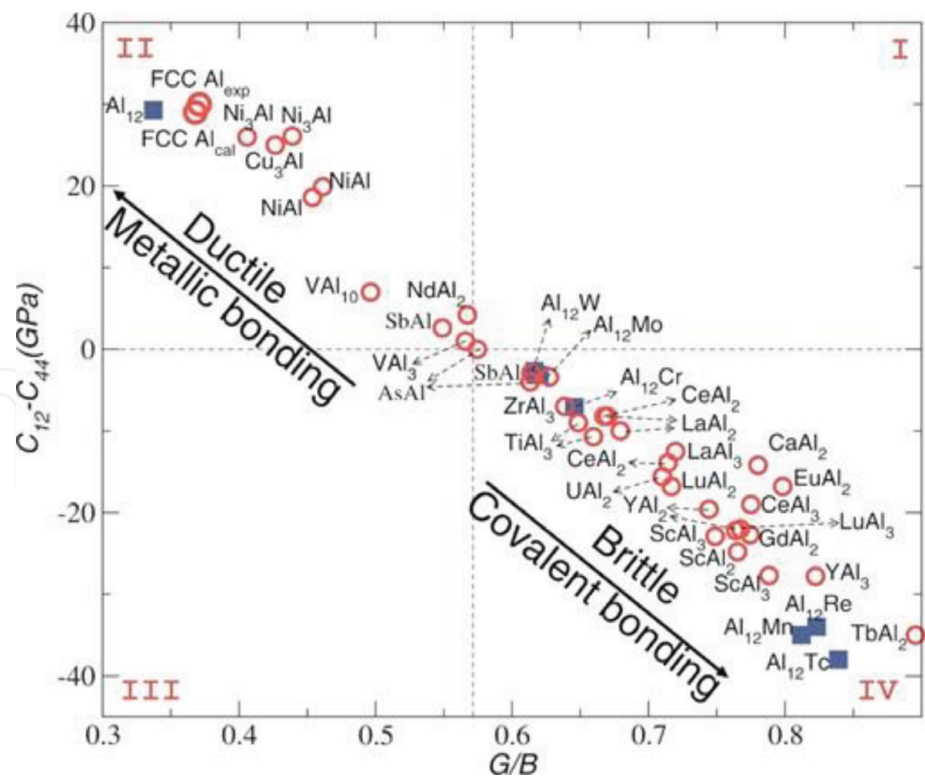


Figure 2.
Correlation between $C_{12}-C_{44}$ and G/B for 35 aluminides (culled from [29]).

for a wide variety of aluminide group of materials is displayed in **Figure 2**. As can be seen, it is evident that an intrinsic correlation between strength and ductility of Al-based materials. It has been observed the criteria indicate a trend in a class of materials with similar deformation mechanism, but is limited by the effects of specimen sizes and crystal structures on deformation processes.

Several authors have studied elastic softening behaviour and recent evidence suggests elastic moduli manifest array of trends with a range of properties including mechanical such as hardness, yield strength, toughness and fragility [22, 30]. In early 1950s, Gilman and Cohen [31] made a historic revelations when they observed that there is a linear correlation between the hardness and elasticity in polycrystalline materials. Nevertheless, successive studies demonstrated that an uniformed linear correlation between hardness and bulk modulus does not really hold for a variety of materials [29] as illustrated in **Figure 3(a)**. Following this, Tester [32] proposed a better empirical link between hardness and shear modulus (G), as illustrated in **Figure 3(b)**. Although, the link between hardness and elastic shear modulus can be arguable, it is certain that he had demonstrated that the shear modulus, the resistance to reversible deformation under shear strain, can correctly provide a key assessment of hardness or ductility criteria for some materials. It is well known that some phase exhibits more hardness or ductility properties than others. Accordingly, it is fair to say that such descriptions could lead to further outlandish discovery in connections with regards to phase components in polycrystalline solids.

4.2 Elastic moduli and martensitic transformation

Martensitic transformation (MT) is a first-order phase type of transformation from a high-symmetry phase (austenite) at high temperature to a crystallographically low-symmetry phase (martensite) at low temperature. Martensitic behaviour has been extensively studied for decades because of its importance

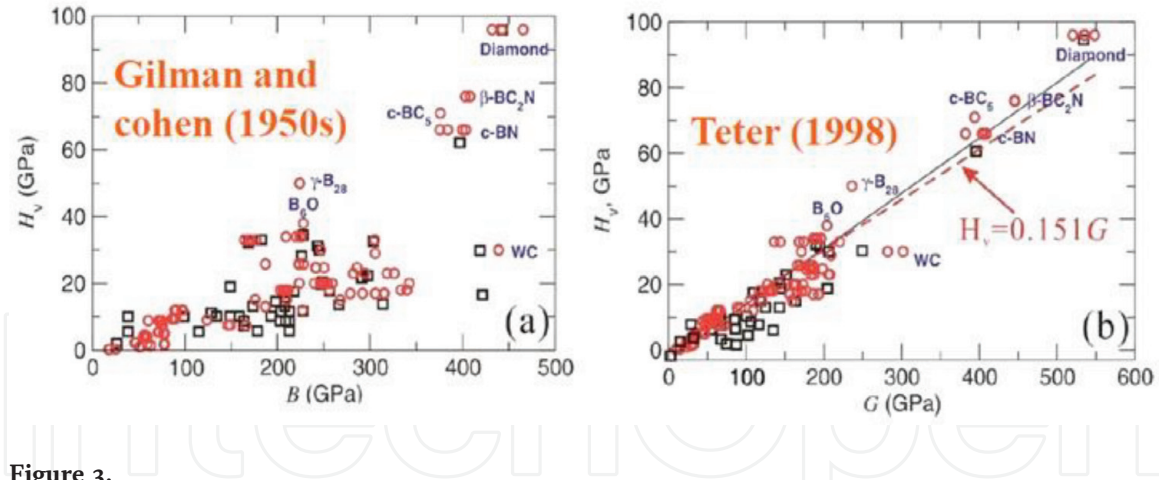


Figure 3. Correlation of experimental Vickers hardness (H_v) with (a) bulk modulus (B) and with (b) shear modulus (G) for 39 compounds [29].

in metallurgy and its key role in shape memory phenomenon. Shape memory alloys (SMA) are materials such as TiNi and TiNi-based alloys [33], Ti-Nb [34], Ti-Mo [20, 31] etc. that exhibit diffusion-less first-order martensitic phase transitions induced by the change of temperature and/or stress. The relation between softening of elastic constants and martensitic transformation has attracted considerable attention for many years and has been discussed by many researchers [35, 36]. This interesting feature of martensitic transformation in shape memory alloys is the existence of precursor phenomena [1, 2]. The relations between MT temperature and elastic constants were investigated by Ren et al. [36]. Experiments [37] indicate that martensitic transition occurs at almost constant values of C' . Slight change in composition would cause strong deviation in the critical temperature at which C' softens to a critical value and martensitic transition occurs. In some alloys exhibiting martensitic transformation, softening of elastic constants $C' = (C_{11} - C_{12})/2$ and large elastic anisotropy, $A = (C_{44})/C'$ was observed in the parent phase, but the significance of the softening is largely different between the alloys. For example, Earlier Takashi Fukuda and co-workers [34] observed the value of C' near the transformation start temperature is approximately 0.01 GPa in In-27Ti (at %) alloy [38], 1 GPa in Au-30Cu-47Zn (at %) alloy [37], 5 GPa in Fe-30Pd (at %) alloy [39], 8 GPa in Cu-14Al-4Ni (at %) alloy [9], and 14 GPa in Ti-50.8Ni (at %) [33] and Al-63.2Ni (at %) alloys [40]. Because of such a large distribution of C' at the M_s temperature, the influence of softening of C' on martensitic transformation is expected to be significantly different between these alloys. Martensitic transformation in some alloys is probably strongly related to the softening of C' , while that in others is weakly related despite the fact that the softening appears before the transformation.

Previously, Zener [5] established a correlation between the magnitudes of $C' = (C_{11} - C_{12})/2$ elastic shear modulus in metallic bcc structures with the occurrence of martensitic phase transformations suggesting links with phase stability, via the atomic interactions. He observed that the large value suggests that C' is much smaller than C_{44} and that MT temperature is dominated by C' [5]. Thus, independent elastic constants are needed to characterize the material response, such as Martensitic transformations (MTs), Shape memory etc. Martensitic transformations (MTs) are often accompanied by elastic modulus softening (acoustic phonon softening) [5]. This explains the strong

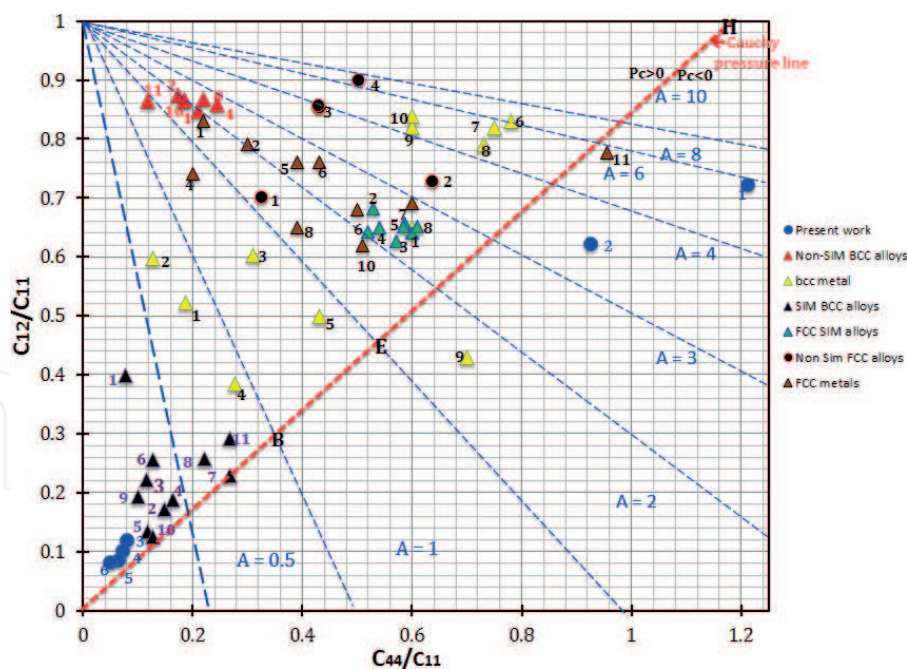


Figure 4. Correlations between parameters reduced elastic-stiffness coefficients $(C_{12})/(C_{11})$ vs. $(C_{44})/(C_{11})$ for several classes of shape memory materials (culled from [34]).

composition-dependence of MT temperature. As a result, the modulus softens abruptly within a narrow temperature window around martensitic start temperature, M_s . However, this is unsurprising since it is well known that they are a consequence of weak restoring forces in specific crystallographic directions that announce the possibility of a dynamical instability. The elastic constants are closely related to the acoustic lattice vibrations or even atomic bondings in crystals, and accordingly will be related to the transformation mechanism for not only the Martensitic alloys but also any other compounds which accompany shear-like or displacive transitions.

Following the above, Nnamchi et al. [41] in a recent study considered the link between different groups of shape memory materials with elastic systematics found a clear delineated in a 2D plot of two dimensionless ratios of elastic constants or reduced elastic-stiffness coefficients, $(C_{12})/(C_{11})$ vs. $(C_{44})/(C_{11})$ formally popularised earlier by Blackman [42], It is only one table with different sections. (see **Figure 4** and **Tables 1** and **2**). This reveals among others the elastic anisotropy, proximity to Born mechanical instability, elastic-constants (interatomic-bonding) changes caused by alloying, pressure, temperature, phase transformations and similarities in types of interatomic bonding. The significance of the softening is largely different between the alloys. Inspecting the diagram, we notice materials with similar chemical bonding tend to fall in the same region of the diagrams. Such diagrams provide many uses.

	This work	C_{11}	C_{12}	C_{12}/C_{11}	C_{44}/C_{11}	Ref.
1	Ti-3Mo	159.3	115	0.72	1.21	[34]
2	Ti-6Mo	111.3	69.07	0.62	0.93	[34]
3	Ti-10Mo	167	19.6	0.12	0.081	[34]
4	Ti-14Mo	179.2	17.9	0.10	0.074	[34]

	This work	C ₁₁	C ₁₂	C ₁₂ /C ₁₁	C ₄₄ /C ₁₁	Ref.
5	Ti-18Mo	192.6	16.3	0.085	0.066	[34]
6	Ti-23Mo	197.5	16	0.081	0.051	[34]
Non-SIM (BCC) alloys						
1	Ti50Ni30Cu20	209	183	0.88	0.17	[43]
2	Ti-50Ni	165	140	0.85	0.21	[44]
3	Ti-29Nb-13Ta-4.6Zr	67.1	39.9	0.87	0.19	[45]
4	Ti-30Nb-10Ta-5Zr	128	92	0.86	0.24	[46]
5	Ti-35Nb	163.5	142	0.87	0.22	[47]
6	Ti-30Nb-5Ta-5Zr	70	30	0.87	0.185	[48]
7	Ti-32.7Nb-11.6Ta-4.49Zr-0.066O-0.052N	137	91.1	0.86	0.12	[49]
Non-SIM (FCC) alloys						
1	Ag-75Au	230	161.5	0.702	0.33	[43]
2	Cu-4.17Si	117	85.2	0.73	0.64	[43]
3	α-Ag-2.4Zn	190	162	0.85	0.43	[43]
4	α-Cu-9.98Al	199	179	0.89	0.50	[43]
5	α-Cu-22.7Zn	158.9	136.2	0.86	0.43	[43]
SIM (BCC) alloys						
1	Ti-35.37Nb	130.2	52	0.40	0.078	[50]
2	Ti-35Nb-2Zr-0.7Ta	183	31.4	0.17	0.15	[49]
3	Ti-35.4Nb-1.9Ta-2.8Zr-0.37O	122	27	0.22	0.11	[49]
4	Ti-24.1Nb-4Zr-8.06Sn-0.15O	140	26.3	0.19	0.16	[49]
5	Ti-35Nb-10Ta-4.6Zr-0.16O	102.5	36	0.16	0.12	[51]
6	Ti-23.9Nb-3.75Zr-8.01Sn-0.04O	157.2	36	0.26	0.127	[51]
7	Ti-24Nb-4Zr-7.9Sn-0.17O			0.23	0.22	[51]
8	Ti-24Nb-4Zr-7.6Sn-0.07O	122	31.4	0.26	0.21	[49]
9	Ti-35.2Nb-10.5Ta-4.97Zr-0.091O-0.014N	140	27	0.19	0.1	[51]
10	Ti-23.9Nb-3.8Zr-7.61Sn-0.08O	102.5	26.3	0.12	0.13	[51]
11	Ti-24Nb-4Zr-7.9Sn	157.2	46	0.29	0.27	[52]
SIM (FCC) alloys						
1	Cu44.9- 50Zn	125	80	0.64	0.6	[43]
2	Au47.5-50Cd	142	96.77	0.68	0.53	[43]
3	Ag45-50Zn	132.8	83.16	0.63	0.57	[43]
4	γ-FeNi	209	183	0.65	0.54	[53]
5	CuAlNi	142.8	93.7	0.66	0.59	[54]
6	B2-NiTi	162	104	0.64	0.52	[55]
7	Cu2.726Al11.122Ni 0.152	137	89.2	0.65	0.59	[56]
8	Cu2.742Al11.105Ni0.152	136	81.763	0.65	0.61	[56]

Table 1.
Elastic constant of some bcc and fcc metals and alloys.

	BCC elements	C12/C11	C44/C114	Ref.
1	V	0.52	0.19	[53]
2	Nb	0.59	0.13	[53]
3	Ta	0.60	0.31	[53]
4	Mo	0.38	0.28	[53]
5	W	0.5	0.43	[53]
6	Li	0.83	0.78	[53]
7	Na	0.82	0.75	[53]
8	K	0.79	0.73	[53]
9	Ba	0.43	0.7	[53]
	FCC elements			[53]
1	Au	0.83	0.22	[53]
2	Pd	0.79	0.3	[53]
3	Pt	0.74	0.2	[53]
4	Ag	0.76	0.39	[53]
5	Cu	0.76	0.43	[53]
6	β -Co	0.69	0.6	[53]
7	α -Sr	0.65	0.39	[53]
8	γ -Fe	0.68	0.5	[53]
9	Ni	0.62	0.51	[53]
10	δ -Pu	0.78	0.96	[53]

Table 2.
Elastic constant of some bcc and fcc metals and alloys.

5. Summary and future challenges

The following bullet points summarise some of the main challenges facing the community.

- Some empirical elastic relationship such as a low G/B ratio (or high ν) favours toughness but also indicates a fragility in polycrystalline materials, though they can be typically difficult to vitrify in some polycrystalline materials.
- Some empirical correlations exist in most of the metallic elements in the periodic table have been found, and alloy development has moved beyond the bucket chemistry type approach used in the early days of elastic properties research. While a number of general guidelines exist for explaining elastic systematics property formation (such as Zener, and Burger's rules), Pugh and Pettifor's criterion [16, 17] in addition to Blackmans have gone beyond simply stating the chemical species that should be present, and their rough proportions, and instead gives exact elastic relationship. However, a more rigorous that delineated the phase stability using systematics could be envisaged in new future.

Glossary of symbols

Symbols used symbols derived from disambiguation (e.g. d for	
C_{ijkl}	the local elastic constant tensor with $\langle \sigma_{ij}(\mathbf{r}) \rangle$ and $\langle \epsilon_{kl}(\mathbf{r}) \rangle$ as the local stress and strain field at a point \mathbf{r} , respectively, and the angular brackets denote ensemble averages
C_{44}	single crystal bulk modulus; $B^o = (C_{11}^0 + 2C_{12}^0)/3$
$C' = (C_{11} - C_{12})/2$	tetragonal shear modulus
$C_{44}, \mu^* = C_{44}^* = \mu^0 + \frac{\langle \tau_{44} \rangle}{1 + G_{44} \tau_{44}}$	trigonal shear modulus
G	the ratio of shearing stress τ to shearing strain γ within the proportional limit of a material
B	bulk modulus, ratio between the fluid pressure and the Volumetric Strain
E	modulus of elasticity or Young's modulus
G	modulus of rigidity or shear modulus
V_L and V_S	the ultrasonic longitudinal and shear wave velocities respectively
ρ	the density of the material
$A = (C_{44})/C'$	elastic anisotropy
U	the energy of the crystal, and quadratic function of the strains
V_0	equilibrium volume
e	an elastic strain
$\langle \sigma_{ij}(\mathbf{r}) \rangle$	effective stiffness of C_{ijkl}
$\epsilon = \epsilon^0 + GT\epsilon^0$	ϵ^0 and GT are the strain and modify Green's function
T	T-matrix is given by $T = \delta C(1 - G\delta C)^{-1}$
I	equivalent to the unit tensor
\tilde{Y}_*	the homogenised polycrystalline Young's modulus
$\tilde{\mu}^*$	homogenised polycrystalline Poisson's ratio
M_S	martensite formation start temperature
M_F	martensite finish temperature
SME	shape memory effect

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